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CHEMISTRY INCLUDING METALLURGY SERIES No. 2

OLAVI ERÄMETSÄ

**Ion Characteristics. A New Way of Assessing the
Chemical Properties of Ions**

Finnish Contribution No. 3

HELSINKI 1958

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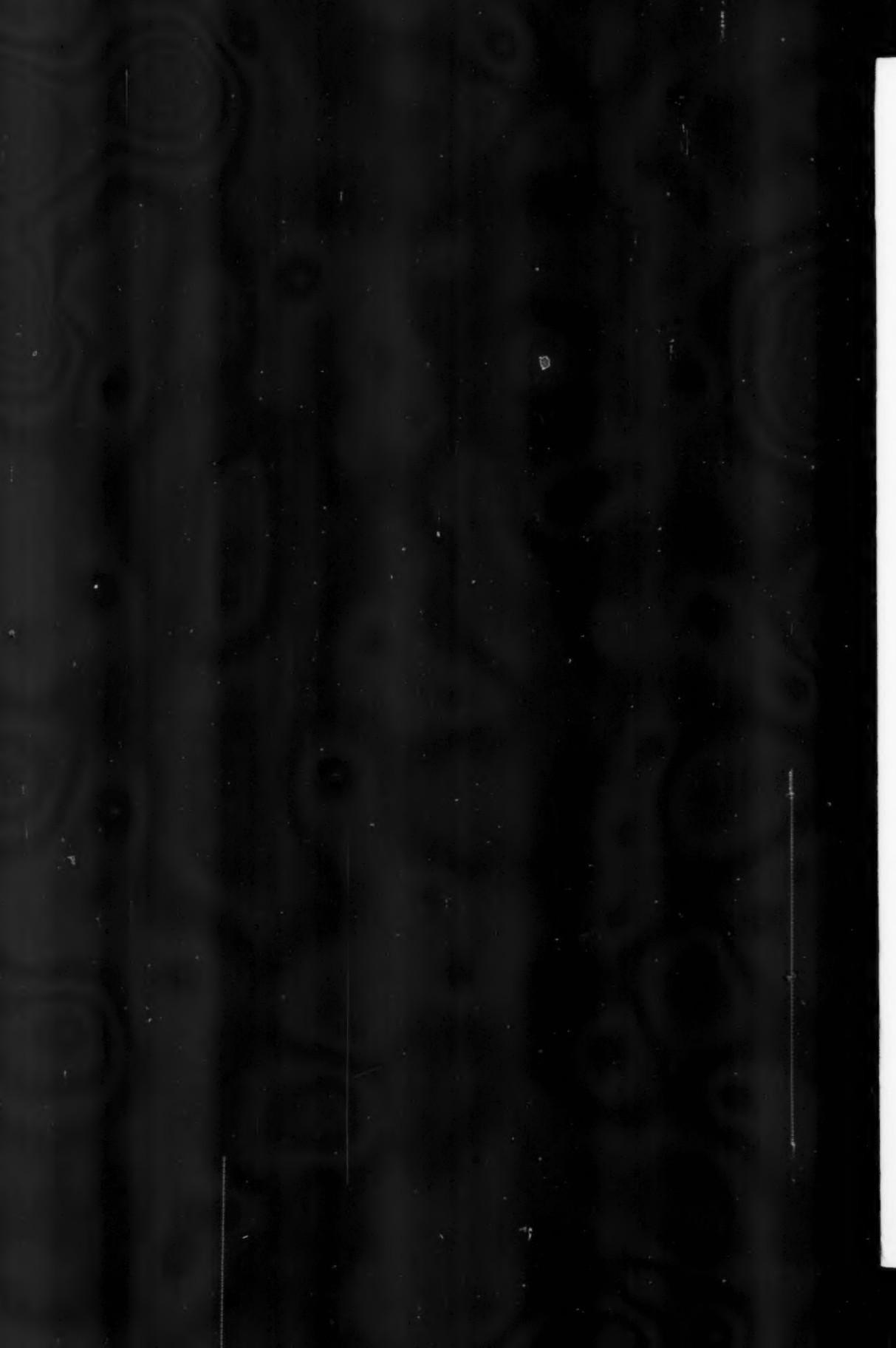
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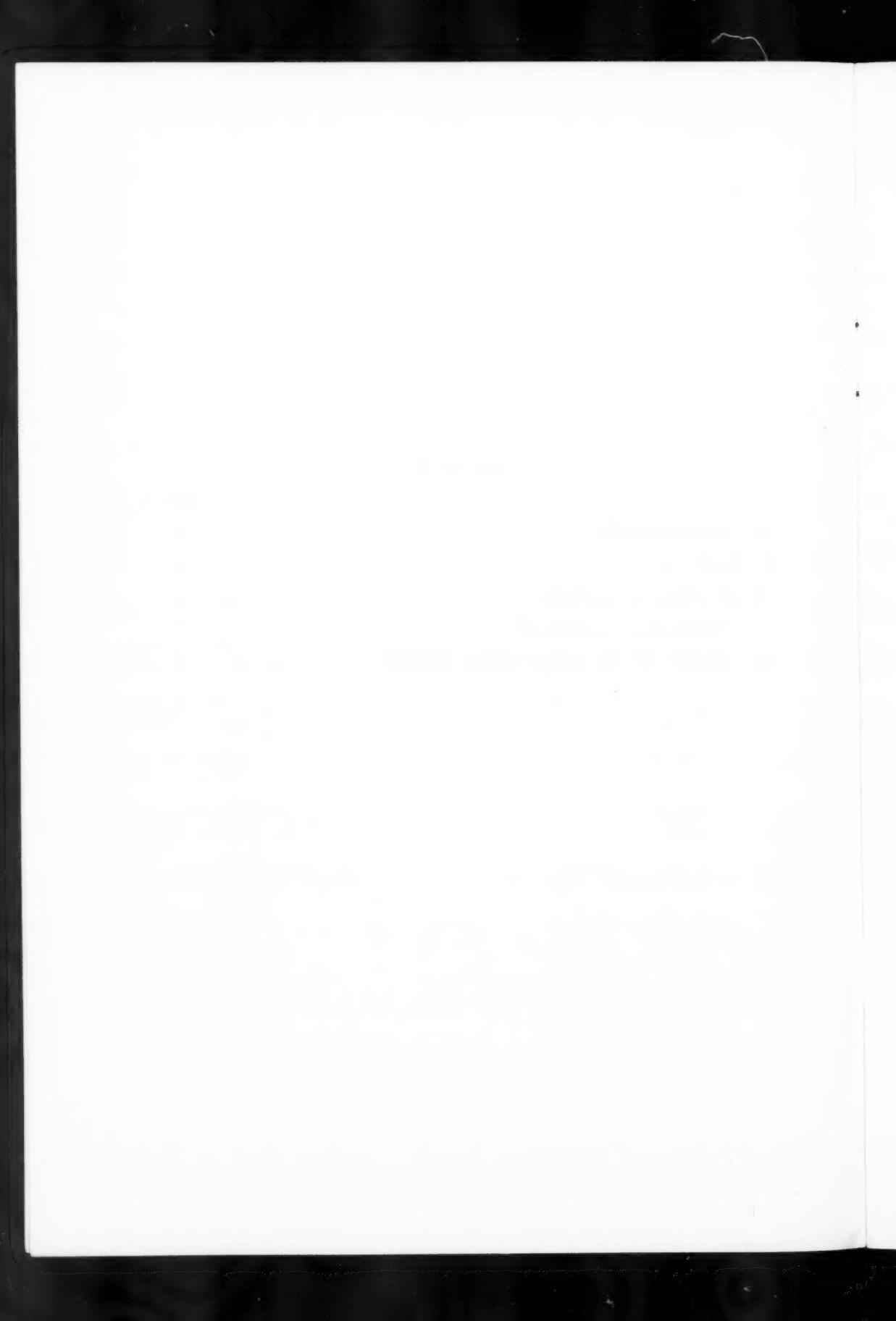
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ION CHARACTERISTICS

By the expression "ion characteristics" - in the following abbreviated IK - is understood the quotient

$$\frac{V^2 + V(Z-V)K_V \cdot e^{-d} \cdot e^{-r_i/r_0}}{r_i}$$

where

V = the nominal ionic charge

r_i = the ionic radius

$r_0 = 0,53 \text{ \AA}$

$K_V = \sin^2(V/4)R$

R = a right angle

e = the base of Neperian logarithms

d = the average surface density of electrons on the atomic subshells, obtained by projecting the electrons into a sphere with a radius equal to the ionic radius.

$$d = \frac{N(e)}{4\pi(r_i/r_0)^2}$$

Only the electrons of the corresponding inert gas should be taken into consideration in the number $N(e)$ of electrons. The screen value of the electrons outside the

sphere of inert gases is 0, the value of the s-, p-, d-electrons inside the sphere (of inert gases) is 1 and the one of f-electrons 1/3.

As the derivation of the formula is based upon certain postulates, the real meaning of which has been inconclusively defined, the formula is postulated as such. Thus it will be treated in the following as an empirical standard characterising the chemical behaviour of ions.

By inserting the known numerical values of the symbols for different ions table I has been obtained. For calculation of the figures in the table Goldschmidt's ionic radii - based upon $O^{2-} = 1,32 \text{ \AA}$ - have been used throughout. The numerical values are as found in "Geochemistry", by K. Rankama and Th. G. Sahama (1949). These ionic radii correspond to a six-co-ordinated structure. As the degree of uncertainty inherent in the ionic radii is greater than 0,01 \AA units, too great a significance cannot be attached to the third figure of the IK. At the end of this paper - in connection with a discussion on the actinide elements - a second table of IK:s is given. This has been calculated assuming that the radius of $O^{2-} = 1,40 \text{ \AA}$.

	r_i	IK		r_i	IK
N^{5+}	0,15	173	Ga^{3+}	0,62	27, 2
S^{6+}	0,34	113	As^{3+}	0,69	26, 0
Se^{6+}	0,35	108	Ce^{4+}	1,02	25, 6
Cr^{6+}	0,35	106	U^{4+}	1,05	25, 0
(C^{4+})	0,18	96, 9)	Co^{3+}	0,65	24, 4
P^{5+}	0,35	81, 5	Th^{4+}	1,10	24, 1
Te^{6+}	0,56	70, 8	Fe^{3+}	0,67	23, 6
As^{5+}	0,47	70, 1	Al^{3+}	0,57	23, 5
V^{5+}	0,4	69, 8	Cr^{3+}	0,64	23, 4
Mo^{6+}	0,62	64, 7	V^{3+}	0,65	22, 8
W^{6+}	0,62	61, 4	Mn^{3+}	0,70	22, 3
Si^{4+}	0,39	52, 3	Ti^{3+}	0,67	21, 3
B^{3+}	0,20	50, 8	Rh^{3+}	0,68	20, 9
Ge^{4+}	0,44	50, 5	Sb^{3+}	0,90	19, 4
Nb^{5+}	0,69	47, 4	In^{3+}	0,92	18, 6
Mn^{4+}	0,52	44, 5	Sc^{3+}	0,83	17, 3
Ta^{5+}	0,69	43, 7	Tl^{3+}	1,05	16, 6
V^{4+}	0,61	39, 6	Lu^{3+}	0,99	16, 3
Ti^{4+}	0,64	37, 6	Yb^{3+}	1,00	16, 3
Sn^{4+}	0,74	35, 8	Tm^{3+}	1,04	15, 6
Te^{4+}	0,89	32, 1	Er^{3+}	1,04	15, 5
Pb^{4+}	0,84	31, 0	Ho^{3+}	1,05	15, 3
Zr^{4+}	0,87	29, 5	Dy^{3+}	1,07	15, 0
Hf^{4+}	0,86	29, 2	Tb^{3+}	1,09	14, 7

	r_i	IK		r_i	IK
Bi^{3+}	1,20	14,6	Hg^{2+}	1,12	6,51
Gd^{3+}	1,11	14,4	Pb^{2+}	1,32	5,41
Y^{3+}	1,06	14,2	Ca^{2+}	1,06	5,37
Eu^{3+}	1,13	14,0	Eu^{2+}	1,24	5,31
Sm^{3+}	1,13	13,9	Sr^{2+}	1,27	4,71
Be^{2+}	0,34	13,9	Ba^{2+}	1,43	4,20
Nd^{3+}	1,15	13,5	Ra^{2+}	1,52	4,16
Pr^{3+}	1,16	13,3	Cu^{1+}	0,96	1,49
Ce^{3+}	1,18	13,1	Li^{1+}	0,78	1,36
La^{3+}	1,22	12,4	Hg^{1+}	1,12	1,33
Ni^{2+}	0,78	9,09	Ag^{1+}	1,13	1,26
Zn^{2+}	0,83	8,74	Na^{1+}	0,98	1,21
Cu^{2+}	0,83	8,60	Au^{1+}	1,37	1,04
Co^{2+}	0,82	8,43	Tl^{1+}	1,49	0,93
Fe^{2+}	0,83	8,18	K^{1+}	1,33	0,88
Mn^{2+}	0,91	7,19	Rb^{1+}	1,49	0,82
Mg^{2+}	0,78	7,17	Cs^{1+}	1,65	0,74
Cd^{2+}	1,03	6,89	$(\text{NH}_4)^+$	1,43	0,73)
Yb^{2+}	1,03	6,64			

Eying through the series, arranged according to descending values of IK it will become apparent that ions with high IK:s form oxygen acids which are the stronger the higher the value of the IK. The acids of elements with IK:s below 65 may usually - as in analytical work - be converted into an insoluble form, by evaporating to dryness in the presence of hydrochloric acid. Figure 1 shows a plot of IK versus the ionic radius r_i . The elements shown below the diagonal line correspond to the aforementioned treatment the better the farther the respective points lie from this line. Next after these elements follow the cations in order of hydrolysis. If obvious exceptions occur these are due to chemical anomalies. Thus, e.g., Sb^{3+} , Bi^{3+} and Pb^{2+} are not precipitated as the normal hydroxides. Generally speaking, the order of hydrolysis cannot always be unambiguously stated. Moreover, the order often depends on the mode of precipitation. E.g.: by hydrolysis with "Urotropine" (hexamethylene tetrammine) Ti^{4+} , Zr^{4+} , Fe^{3+} , Al^{3+} , Cr^{3+} and La^{3+} are precipitated - in that order - whereas Zn^{2+} , Mn^{2+} , Ni^{2+} and Co^{2+} remain in solution, which is to be expected from the magnitude of their respective IK:s. On the other hand, if the precipitation is carried out by dropwise addition of ammonia, Cu^{2+} will be precipitated before La^{3+} . In fractional precipitation with ammonia the above order will be changed and certain ions will move up on the list and the more so the more noble their standard (electrode) potentials.

Alums are formed by tri-valent cations with IK:s between 18 and 30, whereas ions

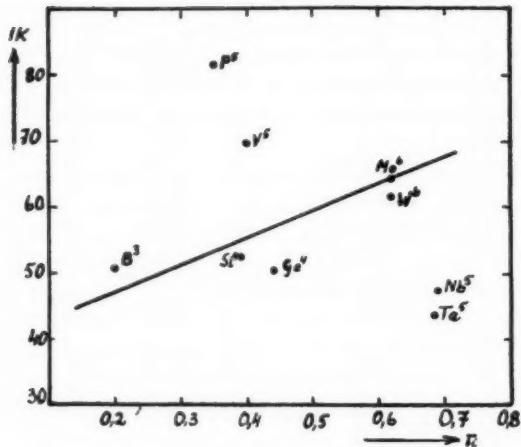


Fig. 1

with lower IK:s do not form alums. The only apparent exception to this rule is Sb^{3+} - which does not form any alums, although it has an IK of 19.4. However, it should be that Sb^{3+} shows an anomalous behaviour in hydrolysis and is capable of existing only in strongly acid solution. In fact, so strongly acid that Fe^{3+} for instance, under similar conditions forms a salt, $Na[Fe(SO_4)_2]$, which is of the same type as the compound formed by tri-valent antimony.

Further, Bi^{3+} occupies a position between the rare earths Tb^{3+} and Gd^{3+} . As is well known, bismuth has been used as a "separating agent" in the fractional crystallisation of rare earth mixtures. In addition the ionic radius of Bi^{3+} is close to those of the rare earths, which makes isomorphism possible. Be^{2+} , on the other hand which has an IK close to those of the cerite rare earth ions, differs completely from these as regards its small ionic radius. Possibly Tl^{3+} could find a use in separations of rare earths.

All cations with IK:s between 4 and 6.7 - Yb^{2+} , Hg^{2+} , Pb^{2+} , Ca^{2+} , Eu^{2+} , Sr^{2+} , Ba^{2+} and Ra^{2+} - form sparingly soluble sulphates. The solubility of the sulphates increases as the upper limit is approached. Thus Yb^{2+} is more difficult to precipitate as the sulphate than Eu^{2+} , for instance.

Sparingly soluble chlorides are formed by all cations with IK:s below 6 - Pb^{2+} , Cu^{+} , Hg^{+} , Ag^{+} , Au^{+} and Tl^{+} - and a standard potential less than +0.5 V. The standard potentials of the other ions in the same range of IK:s are considerably less noble than 2 V, which clearly distinguishes between the two groups. In Figure 2 the IK:s have been plotted against the standard potentials of the respective ions. In the lower left hand corner the region of sparingly soluble chlorides is shown separated by a dotted line. The upper left hand corner contains - well separated from all other

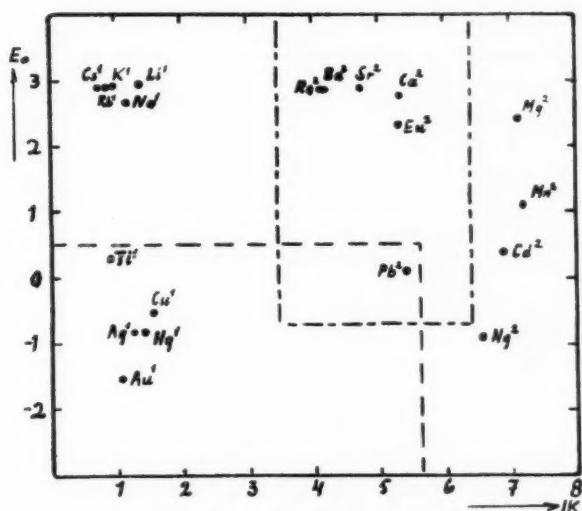


Fig. 2

ions - the alkali metals. The alkaline earth metals, Ca, Sr, Ba and Ra, also form a fairly well defined group, confined within a limited area which, however, borders closely to di-valent ytterbium and, in particular, di-valent europium. Figure 2 shows in addition that the tendency of ions to be precipitated as the sulphides is the stronger the more noble the standard potential, and it shows also that carbonate minerals are formed by ions with IK: between 4 and 13.

In the preceding passages some examples of the importance of the IK have been mentioned. A more detailed account of the usefulness of the IK will be given below where it is considered in connection with other concepts in atomic physics.

IK VERSUS $1/r_i$

In Figure 3 the co-ordinates are IK and $1/r_i$, which both have the same dimensions. The ordinate is shown reduced to an exponential scale. For the sake of clarity only ions with a noble gas configuration have been included, and ions of equal valencies have been connected with lines, drawn from point to point. This gives a Periodic Table arrangement of the elements. However, this arrangement differs from the conventional in that the separation between neighbouring elements is related to each other. In judging the separation between two elements it should be borne in mind that the "a" and "b" groups, respectively, lie in different planes, as will be shown later. Take, e.g., the group Be, Mg, Ca, Sr, Ba and Ra. The distance between the points representing Be^{2+} and Mg^{2+} is very great, in fact, greater than the distance between Be^{2+} and Al^{3+} . Ba^{2+} and Ra^{2+} , on the other hand, lie close together. A similar distribution is found for the alkali metal group. In the series Sc^{3+} , Y^{3+} , La^{3+} - Lu^{3+} the turning row of rare earths is a result of the "lanthanide contraction". The effects of this contraction are felt in the abovelying groups, too. Comparing the close relation between Zr^{4+} and Hf^{4+} it should be remembered that Pb^{4+} - also very close - is situated in an altogether different plane.

Figure 4, a plot of IK versus r_i , corresponds to the lower part of the previous figure. It includes the ions whose electron configuration differs from the noble gas configuration. Several interesting relationships are apparent, e.g., such as Eu^{2+} - Sr^{2+} Pb^{2+} , Th^{4+} - U^{4+} Ce^{4+} , Zn^{2+} - Cu^{2+} - Fe^{2+} - Co^{2+} - Ni^{2+} , etc.

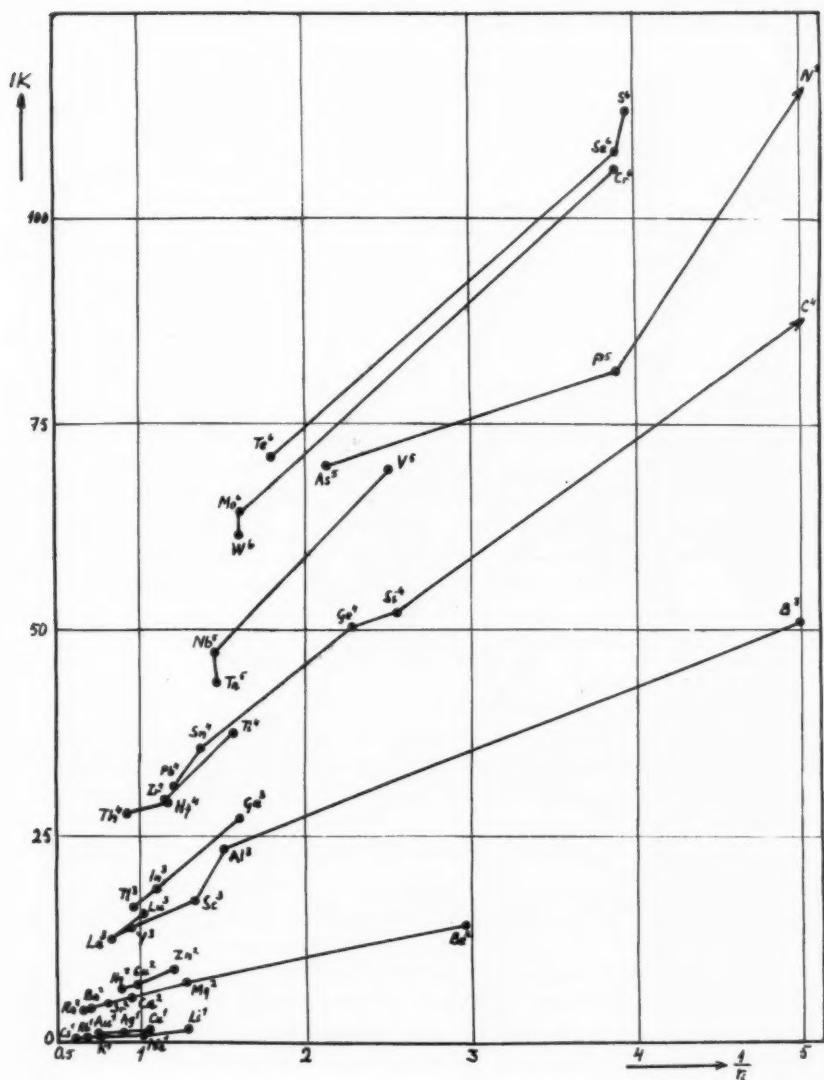


Fig. 3

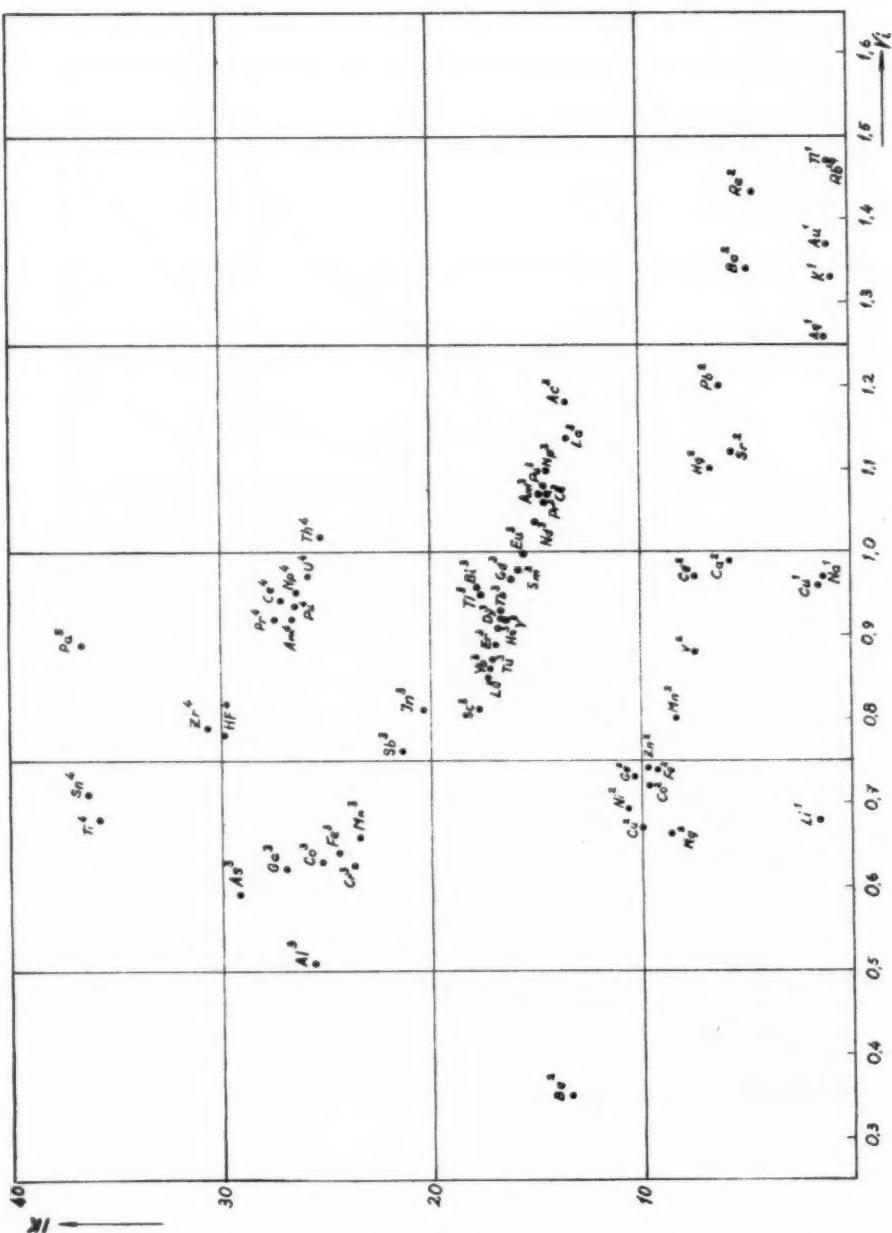


Fig. 4

THE IK VERSUS E_f DIAGRAM

The reduced potential, E_f , is an empirical quantity which is obtained by adding 0.1 volts for each electron outside the filled levels (electron shells) and 0.001 V for every single electron in the ion to the standard electrode potential. As filled levels, or complete configurations, are considered both the noble gas configurations and the (other) filled K, L, M, N, O and P levels. If the number of extraneous electrons is S, the reduced potential is obtained from the equation

$$E_f = E_0 + 0.1 S + 0.001 (Z - V)$$

As this equation results in an obvious over-correction in the case of nickel and cobalt it will be assumed that S cannot exceed 8. In that case S represents the number of the group in the short periodic system, i.e., it equals the "maximum positive valency" of the element concerned.

In Figure 5 the co-ordinates are IK and E_f . For practical reasons IK is reduced to a negative exponential scale. A first glance reveals that the Figure represents the distribution found in "classical" qualitative analysis. The lower left-hand corner contains all cations that are precipitated by hydrochloric acid as sparingly soluble chlorides. However, in the course of conventional analysis copper will not be found in this group, as cuprous copper will be oxidized to cupric copper, shown further to the right. All ions precipitated by hydrogen sulphide from acid solutions are to be found below the horizontal line which corresponds to $E_f = 0.69$. Indium is situated on this line.

The vertical line $IK = 18.7$ divides this region in two. To the left are found the elements that form sulphides insoluble in, to the right those that form sulphides soluble in ammonium sulphide. The value of the ionic radius of di-valent tin is not known with certainty, although interpolation will place stannous tin in the lower

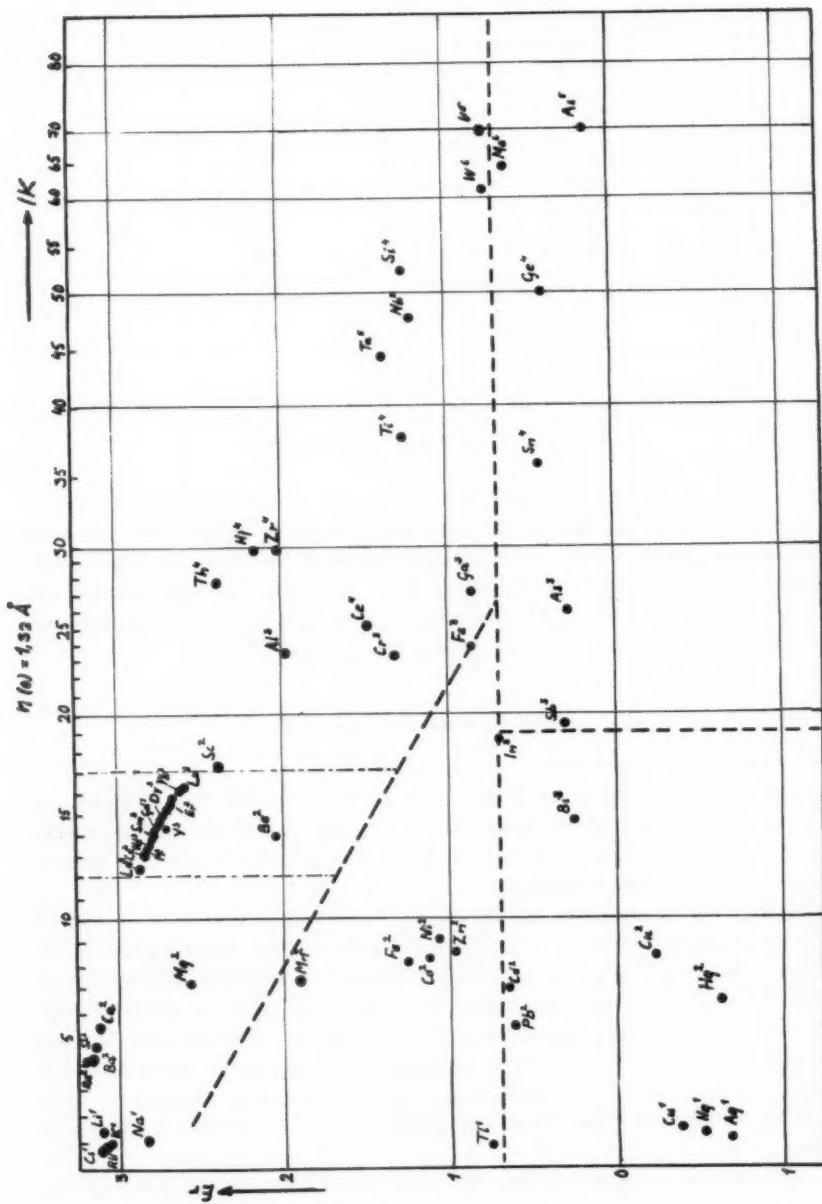


Fig. 5

Kuva 5.

left-hand corner. Stannous sulphide is insoluble in ammonium sulphide but is oxidised by ammonium polysulphide - stannic sulphide, shown in the right-hand region is, of course, soluble in the ammonium sulphides. The tetravalency of the dissolved tin is obvious, since hydrochloric acid precipitates stannic sulphide. What has been said above about tin, applies also to di-valent germanium.

After precipitating the sulphide group any silicic acid present will be converted into an insoluble form - unless this already has been done at an earlier stage - by evaporating to dryness in the presence of hydrochloric acid. Hereby Ta^{5+} , Nb^{5+} , W^{6+} and partly Ti^{4+} , will also be precipitated as insoluble "acids". In the Figure 5 this group is found to the right and above the sulphide region. It is possible to divide the upper region into two by a diagonal line. To the left of this line are to be found all ions that will be precipitated as the sulphides by ammonium sulphide, to the right all those ions that are precipitated as the hydroxides. The line has been drawn through Fe^{3+} which will be precipitated as the sulphide from a cold solution, but as the hydroxide from a boiling solution. If, after removal of the H_2S group, ammonia is added to the solution - containing an excess of ammonium salts - ions with an IK greater than 10 will be precipitated. Using thiosulphate for the hydrolysis will move the limit of the region of precipitation to an IK value of 17. Ferric iron, however, is reduced by the thiosulphate to ferrous iron - IK equal to 8, 18 - and will remain in solution. If ammonium sulphide is added to the filtrate from the ammonia precipitation Zn^{2+} , Ni^{2+} , Co^{2+} , Fe^{2+} and Mn^{2+} will be precipitated as indicated in the figure. Of these only Zn^{2+} will be precipitated from a solution containing acetic acid, as the E_r of this ion is closer to those of the ions in the H_2S group than is the E_r of any of the other ions mentioned above.

Of the remaining elements Mg^{2+} is alone by itself, and Ca^{2+} , Sr^{2+} , Ba^{2+} and Ra^{2+} , as well as the alkali metals form closely spaced groups. Figure 5 also shows some other special features. For instance, all the metals that form complex ammines (Ni, Co, Zn, Cd, Cu) when their hydroxides are treated with an excess of ammonia, are grouped closely together.

At this stage it will be necessary to compare Figure 5 with Figures 3 and 4. In its entity the IK vs E_r vs $\ln r_i$ (or $1/r_i$) diagram forms a three-dimensional "periodic table" - or system of the elements - which can be plotted on paper only by projecting the points representing the ions on to the planes formed by any two of the co-ordinates. Chemical relationship is best studied by comparing the separation between two points in two of the diagrams simultaneously.

The plot of IK vs E_r , in addition to what has been mentioned above also gives an indication of the lithophilic and the chalcophilic properties of the elements. When the IK of an ion is less than 30 the lithophilic, or the chalcophilic properties, respectively, are more pronounced the further away, either above or below, the points representing the ions are situated from the diagonal line. Elements in the vicinity of this line form both sulphide and silicate minerals. When the IK is over 30 the distinction between lithophilic and chalcophilic properties is very slight, or entirely absent. The elements in this group will occur either as free oxides or as anions.

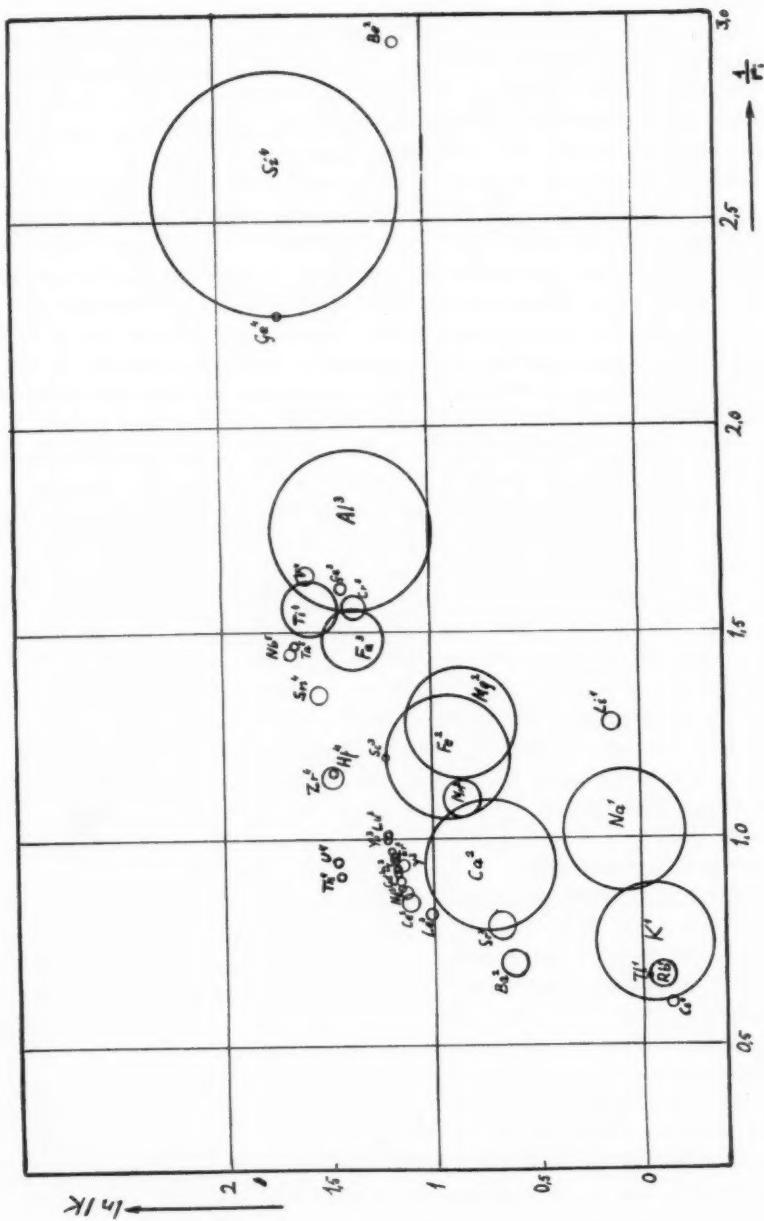


Fig. 6

THE GEOCHEMICAL CAMOUFLAGE

Figure 6 shows a plot of IK vs $\ln r_i$. The radii of the circles representing the ions are proportional to the cubic roots of the abundancies of the elements. In other words, if the circles would be expanded to spheres, the volumes of these spheres would be proportional to the abundancies of the elements in the earths crust. For clarity only the lithophilic elements have been included, as they are all found in almost the same plane.

During the early stages of the crystallisation of a magma minerals with a high melting point are separated. Thereafter minerals are formed by common elements constituting the main part of the magma. Ions shown as small circles inside the larger ones - i.e., elements with an IK and r_i close to that of some of the major elements - remain hidden in the lattices of minerals formed by the major element. For this reason these elements - in spite of the fact that they may be fairly abundant - do not form any minerals on their own. E.g., the abundance of Rb is fifty times that of Cs, but as it is "covered" more completely by potassium than Cs, the latter is capable of forming minerals independently, whereas Rb is not.

From the part of the magma which crystallises last of all, the pegmatites are formed, and as is well known these have long been the best sources of many minerals containing rare elements.

The elements which have become enriched in the pegmatites, are those that lie outside the "main road" (see fig.) formed by the large circles and which have not been "stowed away" in the lattices of the more common minerals of the main crystallisation.

According to Figure 6 Li, Be and B - below the main group of elements - and Sc, Y, La-Lu, Th, U, Nb, Ta, Zr, Hf and Sn, above the same, are to be expected in pegmatites. This agrees well with observed facts.

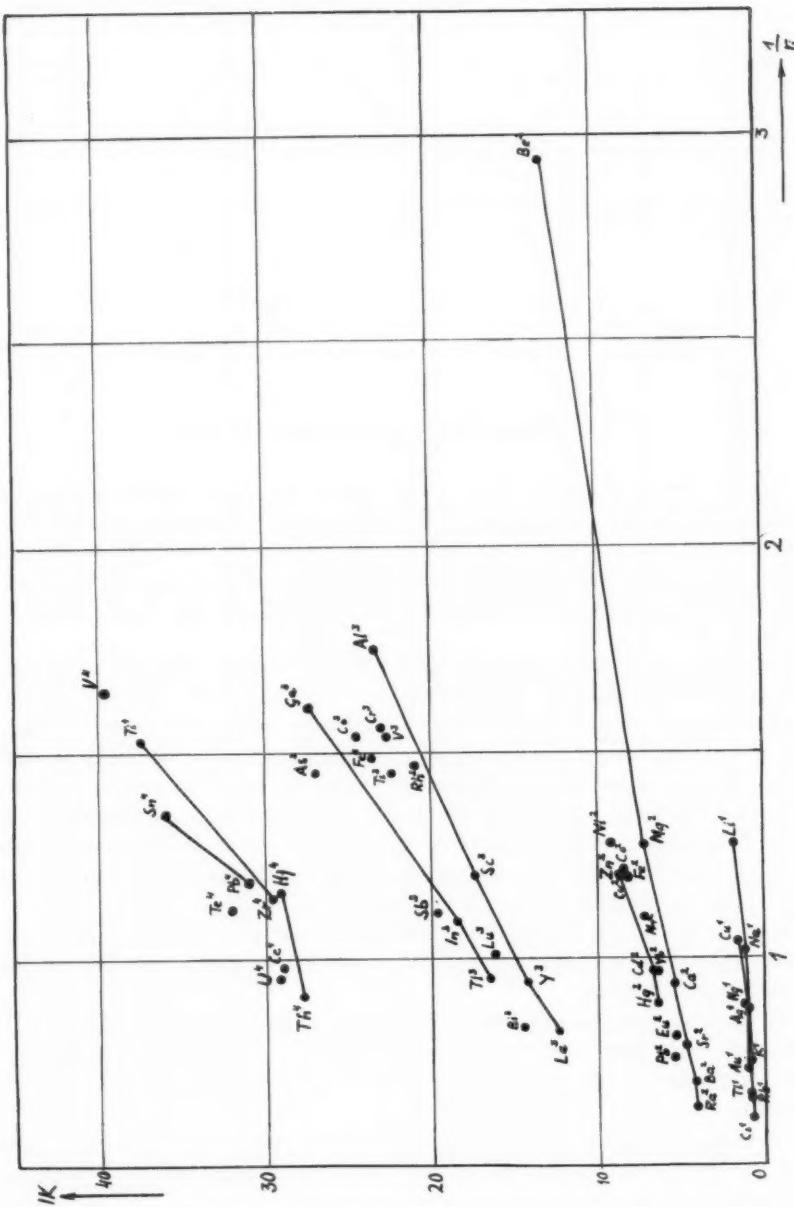


Fig. 7

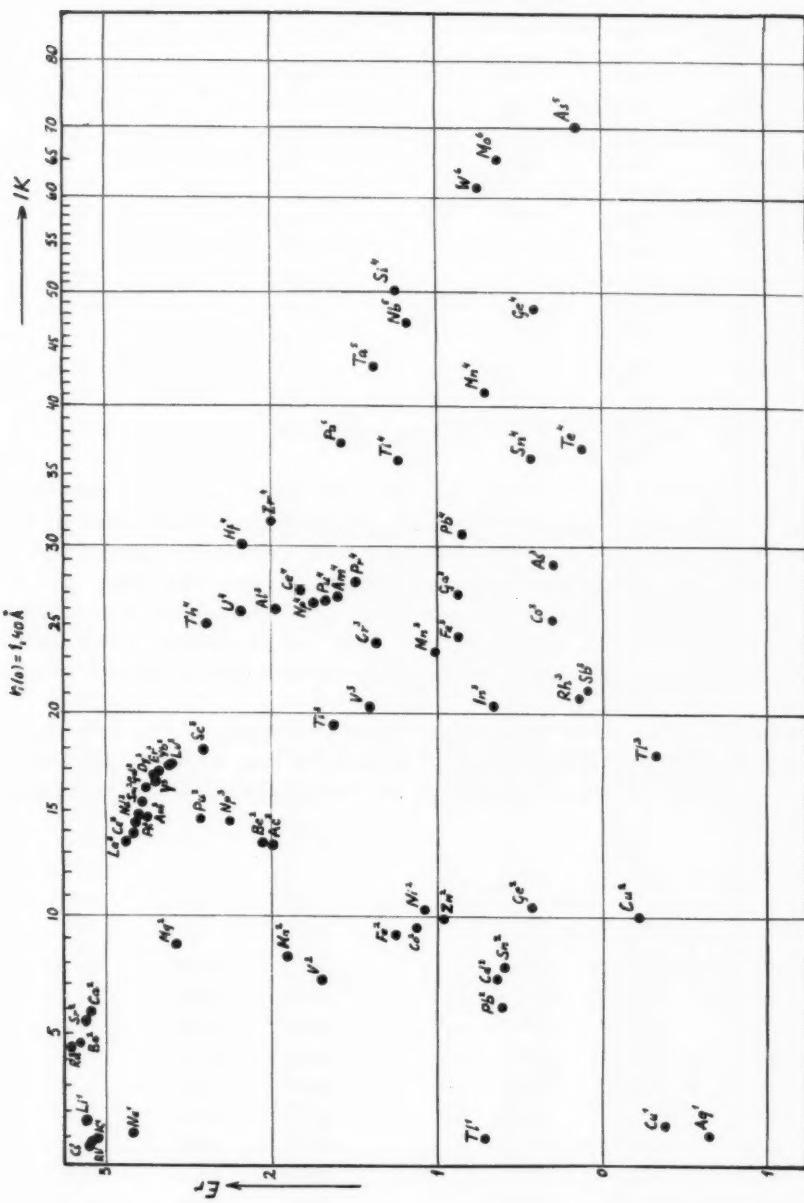


Fig. 8

THE POSITION OF THE TRANSURANIUM ELEMENTS

All the previous IK:s have been calculated from ionic radii based upon $O^{2-} = 1,32 \text{ \AA}$. As the more recently published ionic radii of the transuranium elements have been based upon $O^{2-} = 1,40 \text{ \AA}$, IK:s calculated from these are not directly comparable with those presented above. To make possible a direct comparison all IK:s have been recalculated, using the ionic radii given in a paper by L. H. Ahrens (Geochim. Acta 2, 1952). The table below gives the recalculated values, which differ from those given previously, although the relative order is more or less the same.

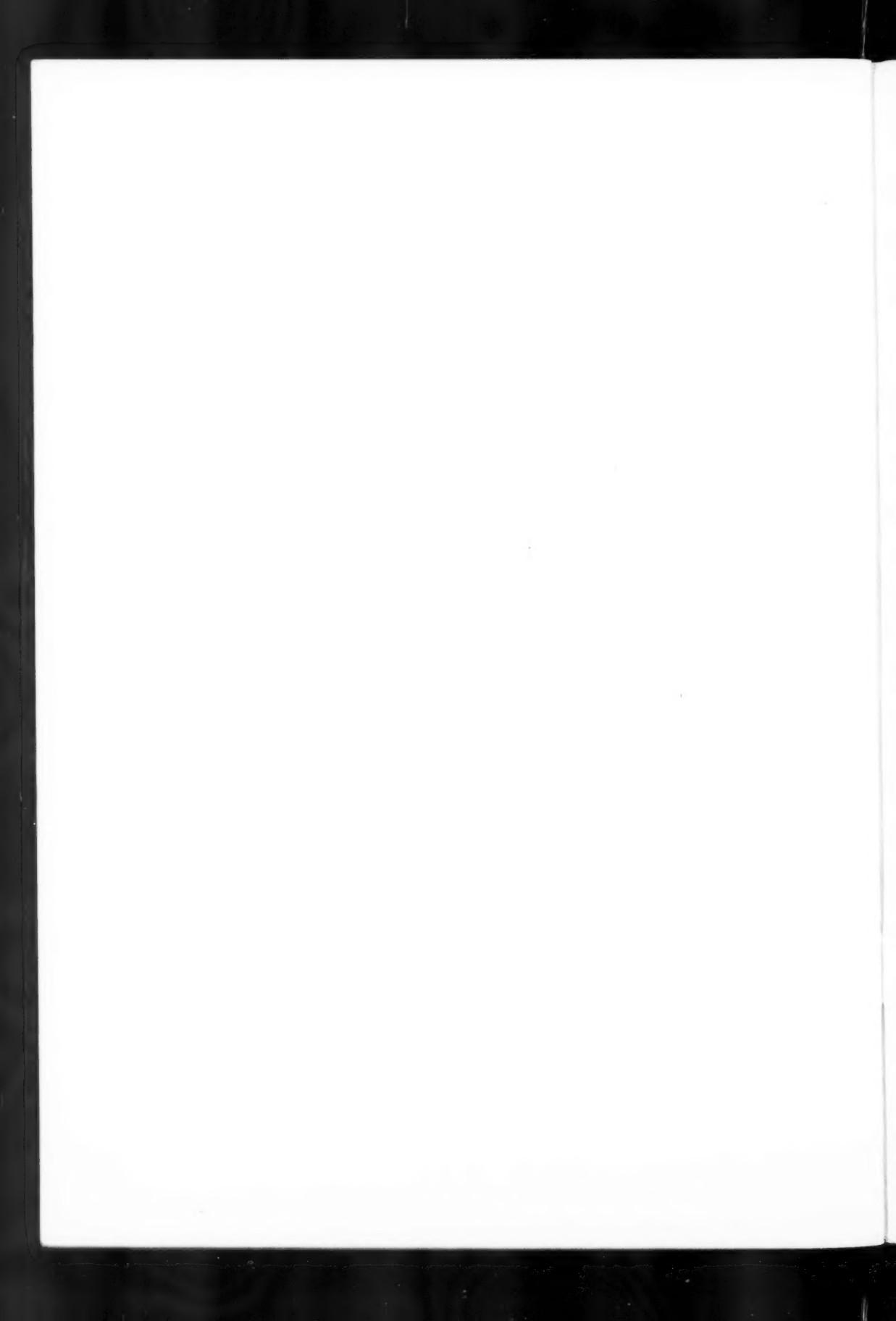
Figure 7 corresponds to Figure 4, and Figure 8 to Figure 5. The similarity of corresponding figures is greater than would be expected from the dissimilarity of the ionic radii. Figure 7 shows unambiguously the chemical nature of the transuranium elements.

	r_i	IK		r_i	IK
N^{5+}	0,13	197	At^{7+}	0,62	80,3
Cl^{7+}	0,27	183	Cr^{6+}	0,52	78,0
S^{6+}	0,30	125	Br^{5+}	0,47	71,7
Mn^{7+}	0,46	109	Te^{6+}	0,56	71,0
C^{4+}	0,16	106	As^{5+}	0,46	70,6
I^{7+}	0,50	99,5	Np^{7+}	0,71	69,9
Se^{6+}	0,42	94,7	Mo^{6+}	0,62	64,8
Tc^{7+}	0,56	89,3	N^{3+}	0,16	64,5
Re^{7+}	0,56	88,2	W^{6+}	0,62	61,5
Cl^{5+}	0,34	85,0	Po^{6+}	0,67	59,0
P^{5+}	0,35	81,5	Os^{6+}	0,69	57,4

	r_i	IK		r_i	IK
S^{4+}	0.37	55, 9	Mn^{3+}	0.66	23, 4
V^{5+}	0.59	55, 8	Sb^{3+}	0.76	21, 4
Cr^{5+}	0.62	53, 0	Rh^{3+}	0.68	20, 9
Sb^{5+}	0.62	52, 6	In^{3+}	0.81	20, 4
Se^{4+}	0.50	50, 6	V^{3+}	0.74	20, 4
Si^{4+}	0.42	50, 3	Ti^{3+}	0.76	19, 4
U^{6+}	0.80	49, 8	Au^{3+}	0.85	18, 0
Ge^{4+}	0.53	48, 8	Sc^{3+}	0.81	17, 8
Nb^{5+}	0.69	47, 4	Bi^{3+}	0.96	17, 8
B^{3+}	0.23	45, 2	Tl^{3+}	0.95	17, 7
Bi^{5+}	0.74	44, 0	Lu^{3+}	0.85	17, 3
Ta^{5+}	0.68	43, 7	Yb^{3+}	0.86	17, 2
Mn^{4+}	0.60	41, 5	Tm^{3+}	0.87	17, 1
V^{4+}	0.63	40, 0	Er^{3+}	0.89	16, 9
Te^{4+}	0.70	37, 0	Ho^{3+}	0.91	16, 8
Pa^{5+}	0.89	36, 6	Dy^{3+}	0.92	16, 7
Sn^{4+}	0.71	36, 2	Tb^{3+}	0.93	16, 6
Pd^{4+}	0.65	35, 9	Y^{3+}	0.92	16, 5
Ti^{4+}	0.68	35, 9	Gd^{3+}	0.97	16, 1
Ru^{4+}	0.67	35, 1	Eu^{3+}	0.98	15, 9
Nb^{4+}	0.74	33, 0	Sm^{3+}	1.00	15, 6
Zr^{4+}	0.79	31, 7	Nd^{3+}	1.04	15, 0
Pb^{4+}	0.84	30, 9	Am^{3+}	1.07	14, 8
Pt^{4+}	0.65	30, 7	Pr^{3+}	1.06	14, 6
Ir^{4+}	0.68	30, 4	Pu^{3+}	1.08	14, 6
Re^{4+}	0.72	30, 3	Np^{3+}	1.10	14, 4
P^{3+}	0.44	30, 0	Ce^{3+}	1.07	14, 4
Hf^{4+}	0.78	29, 9	Be^{2+}	0.35	13, 5
As^{3+}	0.58	29, 2	La^{3+}	1.14	13, 5
Pr^{4+}	0.92	27, 4	Ac^{3+}	1.18	13, 5
Ce^{4+}	0.94	27, 2	Ni^{2+}	0.69	10, 6
Ga^{3+}	0.62	27, 0	Ge^{2+}	0.73	10, 3
Am^{4+}	0.92	26, 7	Cu^{2+}	0.72	10, 0
Pu^{4+}	0.93	26, 5	Zn^{2+}	0.74	9, 90
Np^{4+}	0.95	26, 3	Co^{2+}	0.72	9, 67
Al^{3+}	0.51	25, 7	Fe^{2+}	0.74	9, 25
U^{4+}	0.97	25, 9	Mg^{2+}	0.66	8, 67
Th^{4+}	1.02	25, 2	Pd^{2+}	0.80	8, 45
Co^{3+}	0.63	25, 1	Mn^{2+}	0.80	8, 40
Fe^{3+}	0.64	24, 4	Ag^{2+}	0.89	7, 90
Cr^{3+}	0.63	23, 7	Sn^{2+}	0.33	7, 82

	r_i	IK		r_i	IK
Pt^{2+}	0,80	7,71	Cu^{1+}	0,96	1,49
Cd^{2+}	0,97	7,35	Na^{1+}	0,97	1,22
V^{2+}	0,88	7,25	Ag^{1+}	1,26	1,09
Hg^{2+}	1,10	6,64	Au^{1+}	1,37	1,04
Pb^{2+}	1,20	6,10	Tl^{1+}	1,47	0,95
Ca^{2+}	0,99	5,89	K^{1+}	1,33	0,88
Sr^{2+}	1,12	5,62	Rb^{1+}	1,47	0,84
Ba^{2+}	1,34	4,62	Cs^{1+}	1,67	0,73
Ra^{2+}	1,43	4,54	Fr^{1+}	1,80	0,70
Li^{1+}	0,68	1,58			





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